

Field Suppression of the Density-of-States: A Mechanism for Large Linear Magnetoresistance

D.P. Young, J.F. DiTusa, R.G. Goodrich, J. Anderson, S. Guo, and P.W. Adams

*Department of Physics and Astronomy
Louisiana State University
Baton Rouge, Louisiana, 70803*

Julia Y. Chan
*Department of Chemistry
Louisiana State University
Baton Rouge, LA, 70803*

Donavan Hall
*National High Magnetic Field Laboratory
Florida State University
Tallahassee, FL, 32310
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Hall, resistivity, magnetization, and thermoelectric power measurements were performed on single crystals of the highly anisotropic layered metal LaSb_2 . A 100-fold linear magnetoresistance (MR) was observed in fields up to 45 T, with no indication of saturation. We show that the MR is associated with a magnetic-field-dependent holelike carrier density, $n(H) \propto 1/H$. The effect is orbital, depending upon the component of the magnetic field normal to the layers. At low temperature, a field of 9 T reduces the carrier density by more than an order of magnitude.

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One of the most successful strategies for producing technologically relevant magnetoresistive materials is to enhance the effects of field-dependent magnetic scattering processes through the creation of magnetic superlattices [1] or by doping magnetic insulators such that a magnetic and metal-insulator transition coincide [2]. Unexpectedly, several recent discoveries of a large magnetoresistance (MR) in low carrier density *non-magnetic* metals [4–7] and semiconductors [8] suggest that there may be other ways of realizing field sensitive materials. However, progress to this end has been hampered by the lack of a concise understanding of the physical mechanisms that produce the large linear positive MR found in $\text{Ag}_{2+\delta}\text{Te}$ and $\text{Ag}_{2+\delta}\text{Se}$ [8], LaSb_2 [9], as well as several other non-magnetic materials [3]. In this Letter we show that the large linear MR in LaSb_2 is a consequence of an orbital field suppression of the holelike carrier density, n . The field suppression of n accounts for a linear MR that changes by a factor of 90 by 45 T, with no signs of saturation, suggesting that the system may undergo a field-induced metal-insulator transition at higher fields. The characterization of this density mediated MR may be an important step towards understanding how electronic structure, dimensionality, disorder, and/or correlations give rise to similar linear MR effects in the magnetic rare-earth diantimonide series RSb_2 , and perhaps, other magnetoresistive materials as well.

LaSb_2 is a member of the RSb_2 ($R=\text{La-Nd, Sm}$) family of compounds that all form in the orthorhombic SmSb_2 structure [10,11]. This is a highly anisotropic layered structure in which alternating La/Sb layers and two-dimensional rectangular sheets of Sb atoms are stacked along the c -axis [12]. These structural characteristics give rise to the anisotropic physical properties observed in all the compounds in the RSb_2 series [9]. Here, we have chosen to focus on LaSb_2 , since its low-temperature transport properties are not complicated by magnetic phase transitions which occur in the other members of this series [9].

Single crystals of LaSb_2 were grown from high purity La and Sb by the metallic flux method [13]. The crystals grow as large flat layered plates which are malleable and easily cleaved. The orthorhombic SmSb_2 -structure type was confirmed by single crystal X-ray diffraction. Magnetization was measured with a commercial SQUID magnetometer, and transport properties were performed using a standard 4-probe AC technique at 27 Hz at temperatures from 0.03–300 K and in magnetic fields up to 45 T. Hall effect measurements were made with a 5-wire geometry with data being taken in both positive and negative fields up to 9 T.

The in-plane zero-field electrical resistivity, ρ_{ab} , of single crystals of LaSb_2 was measured from 1.8–300 K and found to be metallic ($d\rho/dT > 0$). The residual resistivity ratio (RRR) was large ($\rho_{ab}(300K)/\rho_{ab}(2K) \approx 70–90$), indicating a high sample quality. The inset of Fig. 1 shows a plot of the in-plane resistivity versus T^2 at zero field. The data are linear, indicating that $\rho_{ab} \propto T^2$ up to 60 K. This T^2 dependence has also been reported in YbSb_2 and is believed

to be a consequence of carrier-phonon scattering on a small cylindrical Fermi surface [14]. The details of the Fermi surface topology of LaSb₂ are not known, but it is indeed likely to be similar to that of YbSb₂ and hence the T^2 behavior. Interestingly, LaSb₂ is a Type I superconductor with a Josephson coupling transition at ~ 0.35 K [15]. In the main panel of Fig. 1 we show the in-plane transverse MR ($H \parallel c$) of LaSb₂ at 2 K. The relative MR is positive and nearly linear above 2 T with a high field slope of $\sim 2 \mu\Omega\text{-cm}/\text{T}$. The effect is also large, as the resistance increases by a factor of 90 from 0 to 45 T and shows no tendency toward saturation. The linearity is unusual in that one expects the classical MR to saturate if the carriers are in closed Fermi surface orbits or increase indefinitely as H^2 for open orbits [16].

In order to characterize the sign and density of the charge carriers, we measured the in-plane Hall resistivity, ρ_H , versus magnetic field for several different temperatures as shown in Fig. 2a. The behavior in ρ_H is characteristically different from what would be expected for the classical Hall effect. The data are not linear in field ($\rho_H \propto H$) but are approximately quadratic, indicating a decreasing holelike carrier density. This effect is temperature dependent, being largest at 2 K and progressively reduced with increasing temperature. There are two other significant features in the data shown in Fig. 2a. First, below 1 T the Hall resistivity at 2 K has a negative slope ($d\rho_H/dH < 0$), and thus, ρ_H passes through zero with increasing field. This is an indication of contributions from both electron and holelike orbits at the Fermi surface, similar to what is observed in the low-temperature magnetotransport of YZn [17]. At fields much higher than the field at which $\rho_H \sim 0$ (0.5 T), the Hall resistivity should be asymptotic to $\rho_H \sim HR_hR_e/(R_h + R_e)$, where R_h and R_e are the hole and electron Hall constants, respectively [18]. Thus, in the high field limit ρ_H is dominated by the majority carriers which are clearly holes in LaSb₂. The second noteworthy feature in the data of Fig. 2a is that the curves for the Hall resistivity all cross at ~ 2 T, indicating that the carrier density is independent of temperature at this field. We note that the Hall mobility, $\mu = \rho_H/(H\rho_{ab})$, is independent of field above ~ 1.5 T and is $\sim 0.1 \text{ m}^2/\text{Vs}$ below 20 K (Fig. 3 inset). Thus, all of the Hall and transport data are in the regime $\omega_c\tau \leq 5$, where ω_c and τ are the cyclotron frequency and relaxation time, respectively. Finally, we have demonstrated that the field dependence of the Hall resistivity depends uniquely on the component of the magnetic field perpendicular to the ab -plane (H_\perp) by performing Hall measurements in tilted field. The inset of Fig. 2a shows the Hall resistivity at 2.5 K and 30 K plotted against H_\perp . The data are essentially identical to those in the main panel of Fig. 2a, indicating that the mechanism responsible for the anomalous Hall resistivity cannot be attributed to an orientation-independent Zeeman splitting. Likewise, the transverse MR with field oriented along the layers is an order of magnitude smaller [9].

Further confirmation of the sign and field dependence of n was obtained from the temperature dependence of the thermoelectric power (TEP), S , as measured by the steady state method (Fig. 2b inset). Like the Hall coefficient, the sign of S is an indicator of the majority carrier sign, and at 300 K the TEP is negative and about an order of magnitude larger than that of most good metals, such as Cu [19]. The data are typical of many low-carrier-density metallic systems and suggest that LaSb₂ is n -type at all temperatures in zero field. The main panel of Fig. 2b shows the TEP plotted versus perpendicular field. At 50 K the TEP is constant in field, maintaining a small negative value ($\sim -4 \mu\text{V}/\text{K}$) even up to 9 T. The lower temperature data, however, show a rather remarkable field dependence. At 10 K and zero field, the TEP is $-1.0 \mu\text{V}/\text{K}$ and then increases approximately linearly in field with a positive slope of about $+1 \mu\text{V}/\text{KT}$. At ~ 2 T, the TEP passes through zero and then remains positive at higher fields. This increase in S above 2 T is consistent with a depletion of the density-of-states with increasing field [20].

It is evident that the non-linear Hall resistance, the field dependent TEP, and the large, positive MR of Fig. 1 are related. Indeed, we have found that both the anomalous Hall resistivity and MR data can be modeled by assuming a single, field-dependent carrier density that varies as $n \sim 1/H$ at high field,

$$n(H) = \frac{n_o}{\sqrt{(1 + (\frac{H}{H_o})^2)}}, \quad (1)$$

where n_o is the apparent carrier density at zero-field, and H_o is a variable parameter which effectively determines a characteristic field scale in the system. The field-dependent Hall resistivity is, $\rho_H(H) = H/en(H)$, where e is the electron charge and $n(H)$ is given by Eq.(1). The solid lines in the main panel and inset of Fig. 2a are fits to the Hall resistivity data using this form, where n_o and H_o of Eq.(1) were varied for the best fit. Values of the variable parameter, H_o , extracted from the fits to ρ_H are plotted as a function of temperature in the inset of Fig. 4 (solid circles). Though H_o saturates below 10 K, at higher temperatures it increases linearly with T . The solid line is a guide to the eye and has a slope 0.24 T/K. This number is approximately equal to $k_B/6\mu_B$, where k_B and μ_B are the Boltzmann and Bohr magneton constants, respectively.

Like the Hall data, the MR of LaSb₂ can also be well described by the carrier density dependence of Eq.(1). The free-electron form of the resistivity is simply, $\rho = 1/e\mu n(H)$, where μ is the carrier mobility. Since the mobility above

1.5 T is only weakly field dependent, the relative MR is well described by $\rho(H)/\rho(0) = n_o/n(H)$. The solid line in Fig. 1 is a least-squares fit to the MR where only H_o in Eq.(1) was varied. The fit captures most of the qualitative behavior of data, including the linear dependence at high field. The H_o values extracted from fits at different temperatures are shown as open circles in the inset of Fig. 4. Though smaller than the values obtained from the Hall data, they have the same high temperature slope, $k_B/6\mu_B$.

Figure 3 shows the temperature dependence of the apparent Hall carrier density, $n_{app} = H/(e\rho_H)$, at several magnetic fields. At low temperature n_{app} is very sensitive to field, with the carrier density saturating below 10 K at all fields. At 2 K, for example, the carrier density at 1 T is reduced by an order of magnitude in a field of 9 T. At 2 T, which is the cross-over field in both the Hall resistivity and TEP, n_{app} is temperature independent. The resistivity of LaSb₂ at 2 T is also temperature independent below 30 K. For fields greater than 2 T, n_{app} decreases with increasing temperature, and for fields less than 2 T, n_{app} increases with increasing temperature. Interestingly, the data in Fig. 3 are reminiscent of ρ vs. T plots of systems displaying a magnetic field induced metal-insulator or superconductor-insulator transition [22]. Indeed, the 2-T curve may represent a separatrix between two low-temperature phases of the system [23]. Clearly the high-field phase appears to be somewhat semiconducting in character in that the carrier density decreases with decreasing temperature. Nevertheless, ρ_{ab} vs. T measurements indicated the system was still metallic at 45 T.

While the results from the magnetotransport measurements outlined above indicate a reduction of the carrier density with increasing field, more compelling evidence to this fact is provided by a thermodynamic measurement of the bulk susceptibility of LaSb₂. The magnetization was observed to be diamagnetic and slightly super-linear with the magnetic field applied along the c -axis [9]. In the analysis that follows we assume that the magnetization arises from two primary contributions: Larmor diamagnetism (of the closed-shell ion cores) and Pauli/Landau paramagnetism of the conduction electrons. By subtracting the purely linear Larmor background from the bulk magnetization data, we were able to isolate the Pauli and Landau contributions which are both proportional to the density of states at the Fermi level, $D(\epsilon_F)$. The main panel of Fig. 4 shows $\chi - \chi_L$ versus field, where χ and χ_L are the bulk and Larmor susceptibilities, respectively. For a first approximation, the Larmor term was set equal to three times the molar susceptibility of a xenon ion [18], i.e. $\chi_L = 3 \times (-43 \times 10^{-6} \text{ cm}^3/\text{mole})$. The data points in Fig. 4 now represent an estimation of the Pauli and Landau contributions to the susceptibility. For a free-electron gas, $D(\epsilon_F) \propto n^{1/3}$, so that using Eq.(1) we have,

$$\chi - \chi_L = \alpha \left[1 + \left(\frac{H}{H_o} \right)^2 \right]^{-\frac{1}{6}}. \quad (2)$$

The solid line in Fig. 4 is a fit to the susceptibility using Eq.(2), where α and H_o were varied for the best fit. Even considering simplifications, such as neglecting band effects (i.e. effective masses), electron-electron interactions, and uncertainties associated with assessing the diamagnetic background, the overall structure of the fit is in good agreement with the susceptibility data and is consistent with the non-linear Hall data of Fig. 2.

In conclusion, we find that the linear MR of LaSb₂ is due to an orbital magnetic field suppression of the density-of-states with a corresponding attenuation of the holelike carrier density that does not saturate up to 45 T. Above 2 T, where the MR becomes linear, we find that the temperature derivative of the carrier density changes from negative to positive, suggesting that the system may undergo a metal-insulator transition at sufficiently high field. A large, linear MR has also been reported in a number of the magnetic rare-earth diantimonides [9], which have the same structure as LaSb₂. It has been conjectured that the MR of these materials is a quantum manifestation of the peculiar Fermi surface topology [21] distinct from a field dependent n . Magnetotransport and de Hass-van Alphen studies under hydrostatic pressure should prove interesting and may help to disentangle electronic structure effects from possible field-modulated localization [24] in these highly anisotropic metals.

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[1] M.N. Baibich *et al.*, Phys. Rev. Lett. **61**, 2472 (1988); R.E. Camlery and R.L. Stamps, J. Phys. Cond. Matter **5**, 3727 (1993).

- [2] S. Jin *et al.*, Science **264**, 413 (1994).
- [3] A.A. Abrikosov, Europhys. Lett., **49** (6), 789 (2000).
- [4] I.M. Lifshits and V.G. Peshanskii, Sov. Phys. JETP, **8**, 875 (1959).
- [5] L.M. Falicov and H. Smith, Phys. Rev. Lett. **29**, 124 (1972).
- [6] D.E. Soule, Phys. Rev. **112**, 698 (1958); J.W. McClure and W.J. Spry, Phys. Rev. **165**, 809 (1968).
- [7] In this case a multi-fold positive, linear, magnetoconductance is seen, V.Yu. Butko, J.F. Ditusa, and P.W. Adams, Phys. Rev. Lett. **85**, 162 (2000); V.Yu. Butko and P.W. Adams, Nature **409**, 161 (2001).
- [8] M. Lee, T.F. Rosenbaum, M.-L. Saboungi, and H.S. Schnyders, Phys. Rev. Lett. **88**, art.no. 066602 (2002); R. Xu, A. Husmann, T.F. Rosenbaum, M.-L. Saboungi, J.E. Enderby, and P.B. Littlewood, Nature **390**, 57 (1997).
- [9] S.L. Bud'ko *et al.*, Phys. Rev. B **57**, 13624 (1998).
- [10] R. Wang and H. Steinfink, Inorg. Chem. **6**, 1685 (1967).
- [11] F. Hullinger, in *Handbook of Physics and Chemistry of Rare Earths*, edited by K.A. Gschneidner and L. Eyring (North-Holland, Amsterdam, 1979), Vol. 4, p. 153.
- [12] X-ray analysis determined the lattice constants to be 0.6319 nm, 0.6173 nm, and 1.857 nm for the *a*, *b*, and *c* crystallographic directions, respectively.
- [13] P.C. Canfield and Z. Fisk, Philos. Mag. B **65**, 1117 (1992).
- [14] N. Sato, T. Kinokiri, T. Komatsubara, and H. Harima, Phys. Rev. B **59**, 4714 (1999).
- [15] D.P. Young *et al.*, in preparation.
- [16] I.M. Lifshits, M.Ya. Azbel, and M.I. Kaganov, /em Electron Theory of Metals (Consultant Bureau, New York, 1973).
- [17] J.-P. Jan *et al.*, Phys. Rev. B **9**, 1377 (1974).
- [18] N.W. Ashcroft and N.D. Mermin, *Solid State Physics* (Saunders, New York, 1976).
- [19] F.J. Blatt, P.A. Schroeder, C.L. Foiles, and D. Greig, *Thermoelectric Power of Metals* (Plenum Press, New York, 1976), p. 5.
- [20] *S* is proportional to the logarithmic derivative of the density-of-states, see Ref. [19].
- [21] A.A. Abrikosov, Phys. Rev. B **60**, 4231 (1999).
- [22] D.B. Haviland, Y. Liu, and A.M. Goldman, Phys. Rev. Lett. **62**, 2180 (1989); A. Yazdani and A. Kapitulnik, Phys. Rev. Lett. **74**, 3037 (1995); E. Bielejek and Wenhai Wu, cond-mat/0108232.
- [23] M.P.A. Fisher, Phys. Rev. Lett. **65**, 923 (1990); S.L. Sondhi *et. al.*, Rev. Mod. Phys. **69**, 315 (1997).
- [24] J.C. Phillips, Phys. Rev. B **46**, 8542 (1992).

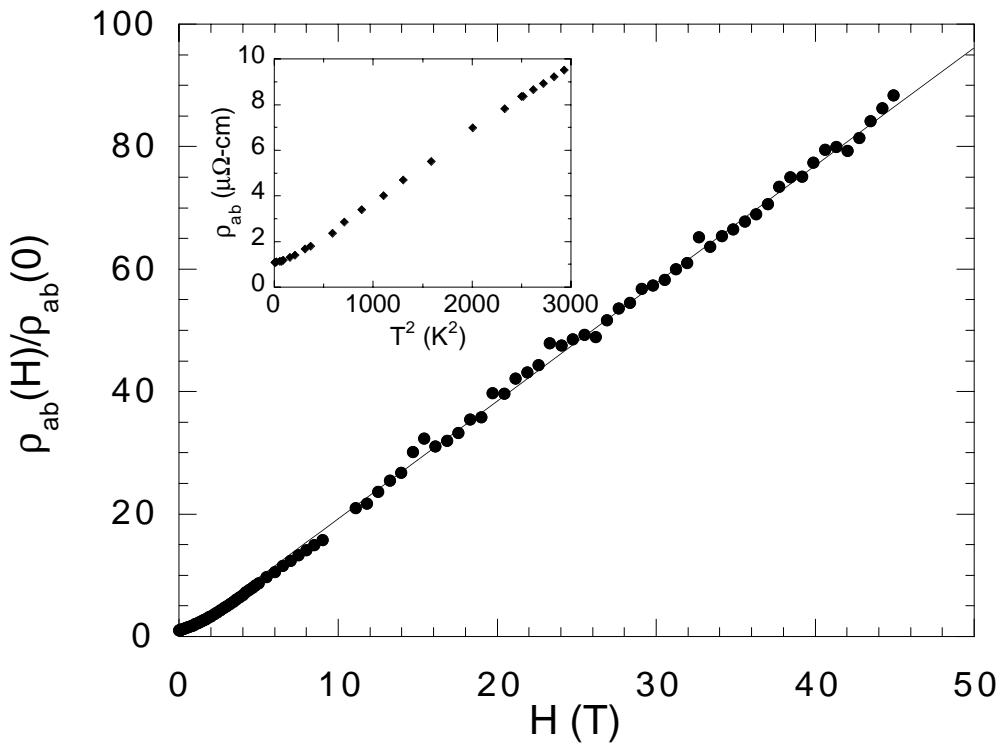


FIG. 1. In-plane resistivity of LaSb₂ at 2 K as a function of perpendicular magnetic field, $\rho_{ab}(0) = 1.0 \mu\Omega\text{-cm}$. The solid line is a fit to the data using Eq.(1) as described in the text. Inset: T^2 temperature dependence in zero field.

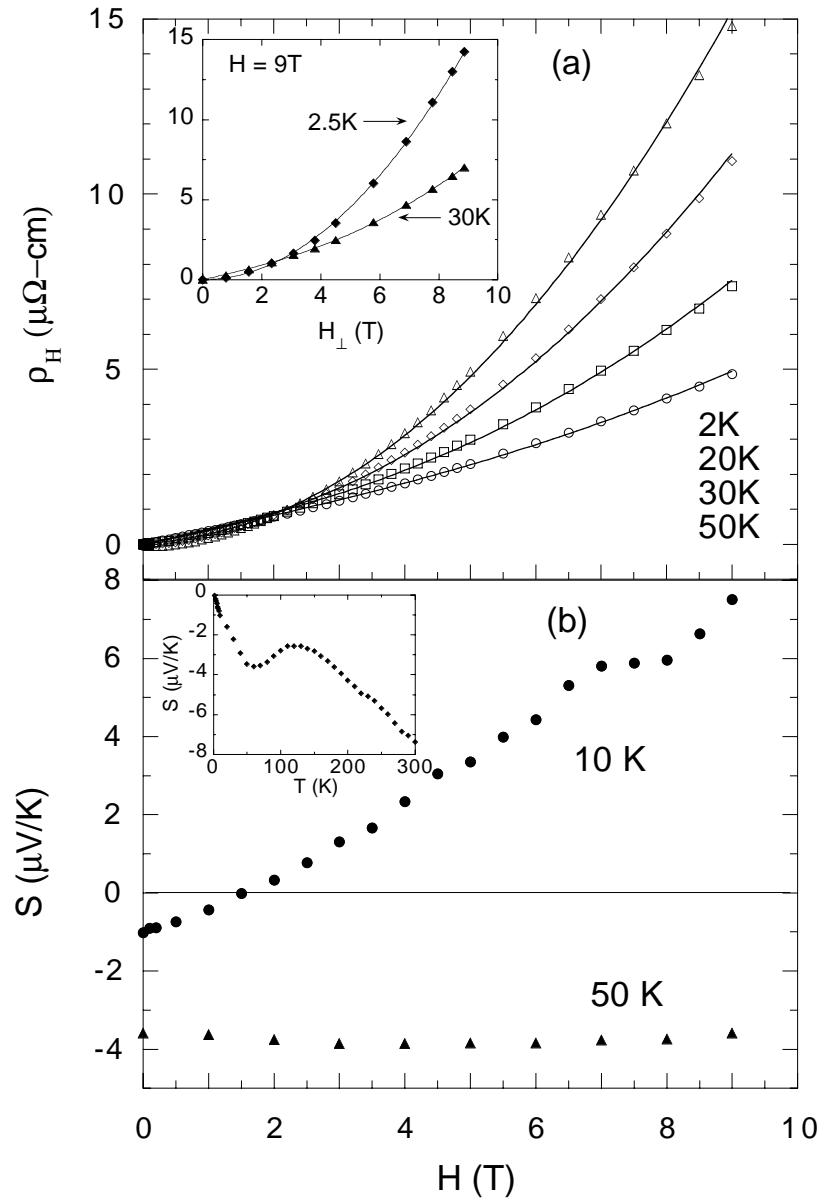


FIG. 2. (a). Hall resistivity as a function of magnetic field oriented perpendicular to the layers. Inset: Hall resistivity obtained by rotation in constant magnetic field, where H_\perp is the field component perpendicular to the layers. The solid lines are fits to the data using Eq.(1) as described in the text. (b) Thermoelectric power as a function of perpendicular magnetic field. Note the sign change at ~ 2 T in the 10 K data. Inset: Temperature dependence of the thermoelectric power in zero field.

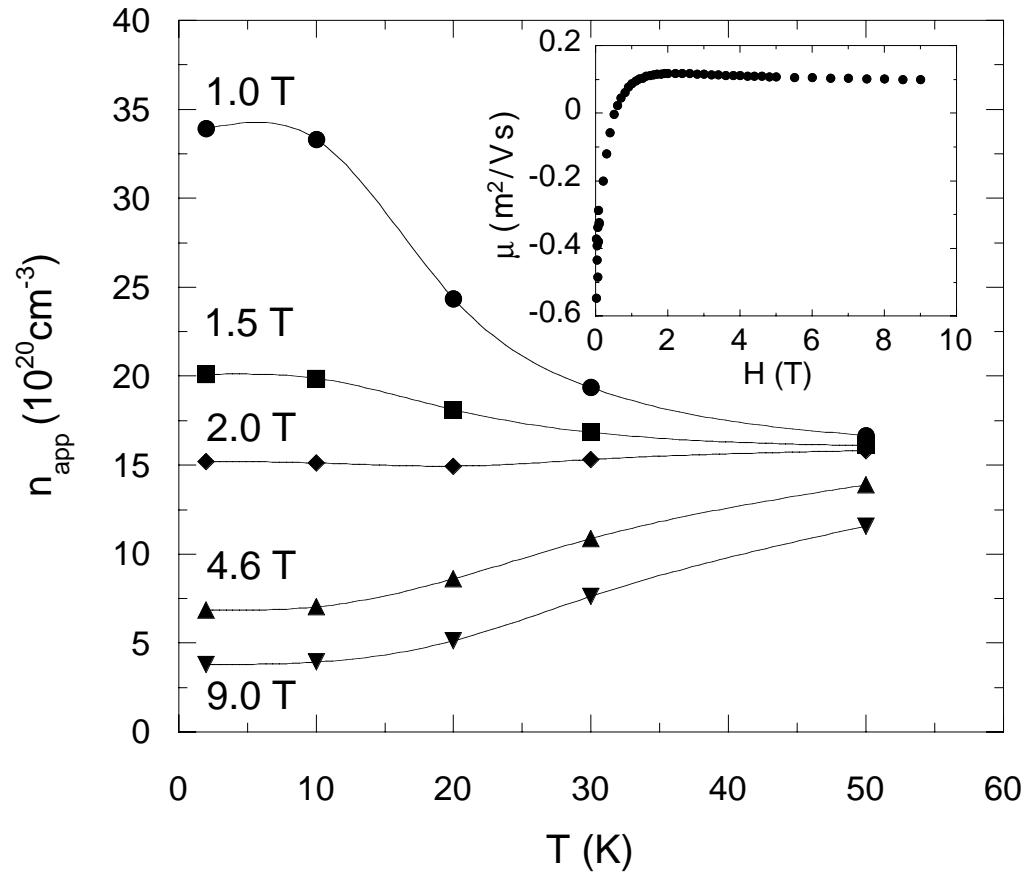


FIG. 3. Temperature dependence of the apparent carrier density as derived from Hall data in Fig. 2. Note that the 2-T carrier density is independent of temperature. The lines are provided as a guide to the eye. Inset: Field dependence of the apparent mobility at 2 K. The large negative values of μ are an artifact of $\rho_H \leq 0$ below 1T.

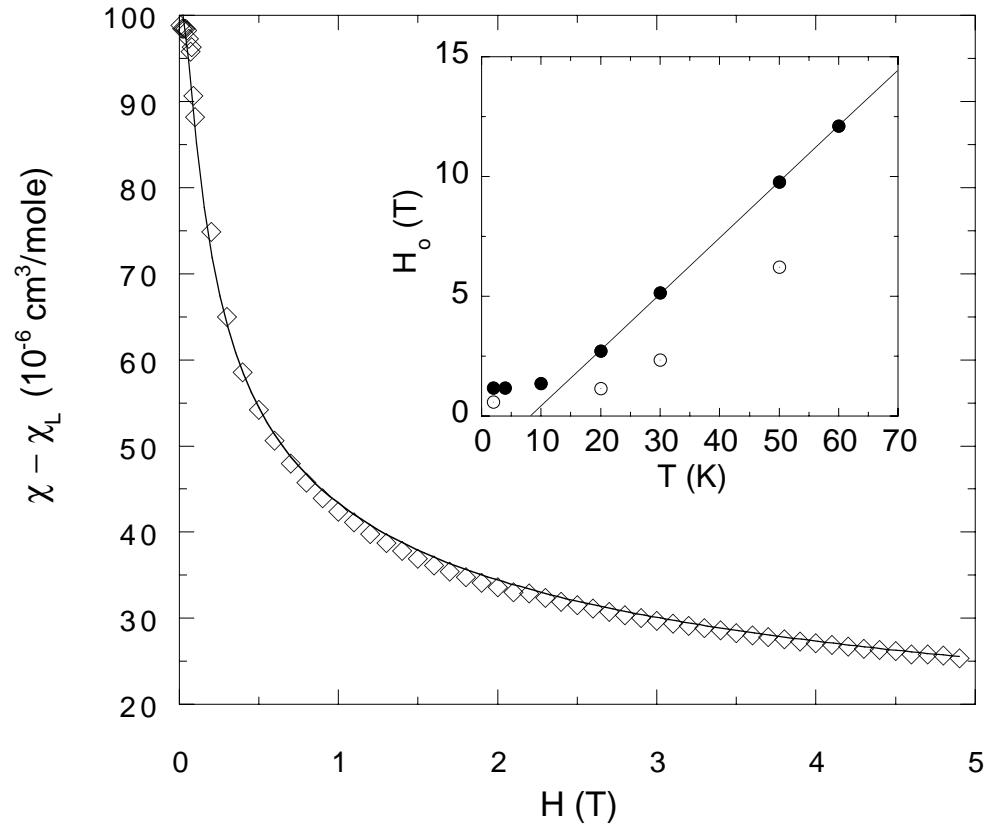


FIG. 4. Susceptibility at 2 K as a function of perpendicular magnetic field. The Larmor susceptibility has been subtracted off leaving behind the Pauli and Landau contributions which are both proportional to the density of states. The solid line is a fit to the data using Eq.(2). Inset: Solid symbols are H_o values obtained from fits to the data in Fig. 2, and the open symbols are values obtained from fits to the relative MR. The solid line is provided as a guide to the eye and has a slope of $k_B/6\mu_B$.